BEST AVAILABLE IMAGES

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PTO/SB/21 (02-04) (AW 02/2004)
Approved for use through 7/31/2006. OMB 0651-0031
U.S. Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE

TRANSMITTAL FORM

(to be used for all correspondence after initial filing)

Total Number of Pages in This Submission 8*

Application Number	10/689,252
Filing Date	October 20, 2003
First Named Inventor	Dharshini C. Fongalland
Art Unit	
Examiner Name	
Attorney Docket No.	JMYT-234US1

ENCLOSURES *(Check all that apply)*

- ☒ Fee Transmittal Form
  - ☒ Fee Attached
- ☐ Amendment/Reply
  - ☐ After Final
  - ☐ Affidavits/Declaration(s)
- ☐ Extension of Time Request
- ☐ Express Abandonment Request
- ☐ Information Disclosure Statement
- ☐ Certified Copy of Priority Document(s)
- ☐ Response to Missing Parts/ Incomplete Application
  - ☐ Response to Missing Parts under 37 CFR 1.52 or 1.53
- ☐ Drawing(s)
- ☐ Licensing-related Papers
- ☐ Petition
- ☐ Petition to Convert to a Provisional Application
- ☐ Power of Attorney, Revocation, Change of Correspondence Address
- ☐ Terminal Disclaimer
- ☐ Request for Refund
- ☐ CD, Number of CD(s) ____
- ☐ After Allowance Communication to Group
- ☐ Appeal Communication to Board of Appeals and Interferences
- ☐ Appeal Communication to Group **(Appeal Notice, Brief, Reply Brief)**
- ☐ Proprietary Information
- ☐ Status Letter
- ☒ Other Enclosure(s) (please identify below):

  Response to Notice of Incomplete Nonprovisional Application

Remarks:

The total number of pages indicated above does not include the total number of pages in the copy of the continuation application as filed.

SIGNATURE OF APPLICANT, ATTORNEY OR AGENT

Firm or Individual Name	Christopher R. Lewis RatnerPrestia	*Registration No. (Attorney/Agent)*	36,201
Signature			
Date	June 1, 2004		

CERTIFICATE OF TRANSMISSION / MAILING

I hereby certify that this correspondence is being facsimile transmitted to the USPTO or deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on this date:

Typed or printed name	Christopher R. Lewis		
Signature		*Date*	June 1, 2004

IFW

PTO/SB/17 (10-03) (AW 12/2003)
Approved for use through 7/31/2006. OMB 0651-0032
U.S. Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE

FEE TRANSMITTAL for FY 2004

Effective 10/01/2003. Patent fees are subject to annual revision.

☐ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT ($) 130

Complete if Known	
Application Number	10/689,252
Filing Date	October 20, 2003
First Named Inventor	Dharshini C. Fongalland
Examiner Name	
Art Unit	
Attorney Docket No.	JMYT-234US1

METHOD OF PAYMENT *(check all that apply)*

☒ Check ☐ Credit Card ☐ Money Order ☐ Other ☐ None

☒ Deposit Account **(use as backup only)**:

Deposit Account Number: 18-0350

Deposit Account Name: RatnerPrestia

The Director is authorized to: *(check all that apply)*
- ☐ Charge fee(s) indicated below
- ☒ Credit any overpayments
- ☒ Charge any additional fee(s) or any underpayment of fee(s)
- ☐ Charge fee(s) indicated below, **except for the filing fee** to the above-identified deposit account.

FEE CALCULATION

1. BASIC FILING FEE

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description	Fee Paid
1001	770	2001	385	Utility filing fee	
1002	340	2002	170	Design filing fee	
1003	530	2003	265	Plant filing fee	
1004	770	2004	385	Reissue filing fee	
1005	160	2005	80	Provisional filling fee	

SUBTOTAL (1) ($) 0

2. EXTRA CLAIM FEES FOR UTILITY AND REISSUE

			Extra Claims		Fee from below		Fee Paid
Total Claims		-20** =	0	X		=	0
Independent Claims		-3** =	0	X		=	0
Multiple Dependent				X		=	0

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description
1202	18	2202	9	Claims in excess of 20
1201	86	2201	43	Independent claims in excess of 3
1203	290	2203	145	Multiple dependent claim, if not paid
1204	86	2204	43	** Reissue independent claims over original patent
1205	18	2205	9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) ($) 0

**or number previously paid, if greater; For Reissues, see above

FEE CALCULATION (continued)

3. ADDITIONAL FEES

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description	Fee Paid
1051	130	2051	65	Surcharge - late filing fee or oath	
1052	50	2052	25	Surcharge - late provisional filing fee or cover sheet.	
1053	130	1053	130	Non-English specification	
1812	2,520	1812	2,520	For filing a request for *ex parte* reexamination	
1804	920*	1804	920*	Requesting publication of SIR prior to Examiner action	
1805	1,840*	1805	1,840*	Requesting publication of SIR after Examiner action	
1251	110	2251	55	Extension for reply within first month	
1252	420	2252	210	Extension for reply within second month	
1253	950	2253	475	Extension for reply within third month	
1254	1,480	2254	740	Extension for reply within fourth month	
1255	2,010	2255	1,005	Extension for reply within fifth month	
1401	330	2401	165	Notice of Appeal	
1402	330	2402	165	Filing a brief in support of an appeal	
1403	290	2403	145	Request for oral hearing	
1451	1,510	1451	1,510	Petition to institute a public use proceeding	
1452	110	2452	55	Petition to revive – unavoidable	
1453	1,330	2453	665	Petition to revive – unintentional	
1501	1,330	2501	665	Utility issue fee (or reissue)	
1502	480	2502	240	Design issue fee	
1503	640	2503	320	Plant issue fee	
1460	130	1460	130	Petitions to the Commissioner	130
1807	50	1807	50	Processing fee under 37 CFR 1.17(q)	
1806	180	1806	180	Submission of Information Disclosure Stmt	
8021	40	8021	40	Recording each patent assignment per property (times number of properties)	
1809	770	2809	385	Filing a submission after final rejection (37 CFR § 1.129(a))	
1810	770	2810	385	For each additional invention to be examined (37 CFR § 1.129(b))	
1801	770	2801	385	Request for Continued Examination (RCE)	
1802	900	1802	900	Request for expedited examination of a design application	
				Other fee (specify)	

*Reduced by Basic Filing Fee Paid SUBTOTAL (3) ($) 130

SUBMITTED BY				Complete *(if applicable)*	
Name (Print/Type)	Christopher R. Lewis	*Registration No. Attorney/Agent)*	36,201	*Telephone*	610-407-0700
Signature				*Date*	June 1, 2004

WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

This collection of information is required by 37 CFR 1.17 and 1.27. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appln. No:	10/689,252
Applicants:	Dharshini C. Fongalland et al.
Filed:	October 20, 2003
Title:	SUBSTRATE
TC/A.U.:	
Examiner:	
Confirmation No.:	3857
Docket No.:	JMYT-234US1

RESPONSE TO NOTICE OF INCOMPLETE NONPROVISIONAL APPLICATION

Mail Stop Petitions
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

Petition for Grant of Filing Date

The Notice of Incomplete Nonprovisional Application (copy enclosed) mailed May 25, 2004, indicates that the specification of the above-identified continuation application is missing, and that a complete specification, accompanied by a newly execution oath or declaration, must be submitted to the U.S. Patent and Trademark Office (PTO) within two (2) months of the date of the Notice.

As this application is a continuation application, a copy of the specification of the parent application, namely, Int'l App. No. PCT/GB99/02935 (WO 00/23510), and a copy of the executed Declaration/Power of Attorney from the parent application, were forwarded to the PTO on October 20, 2003, by Express Mail. A return receipt postcard bearing a sticker from the PTO which includes the filing date and application number of the application, and indicates that these documents were received by the PTO, was received by applicants' representative on October 30, 2003. A copy of the continuation application, as filed, including the specification and the executed Declaration/Power of Attorney from the parent application, as filed on October 20, 2003, are enclosed. Also enclosed is a copy of the return receipt postcard which bears the PTO sticker, and indicates that the aforementioned documents were previously received by the PTO.

The requirements for filing a continuation application were met at the time of filing of this application on October 20, 2003. Therefore, applicants respectfully request that the filing date of October 20, 2003, be granted for this application.

The Petition fee of $130, as set forth in 37 C.F.R. § 1.17(h), is enclosed.

Request for Refund of Petition Fee

As the documents requested in the Notice of Incomplete Nonprovisional Application were submitted to the PTO on October 20, 2003, a refund of the enclosed petition fee is respectfully requested. Please make the refund by returning the enclosed check or by crediting Deposit Account No. 18-0350.

Respectfully submitted,

Christopher R. Lewis, Reg. No. 36,201
Attorney for Applicants

CRL/lrb
Enclosures: Copy of the continuation application, as filed, including the Specification and the executed Declaration/Power of Attorney from the parent application
Copy of the return receipt postcard
Copy of Notice of Incomplete Nonprovisional Application
Check ($130)

Dated: June 1, 2004

P.O. Box 980
Valley Forge, PA 19482-0980
(610) 407-0700

The Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. **18-0350** of any fees associated with this communication.

I hereby certify that this correspondence is being deposited with the United States Postal Service as first class mail, with sufficient postage, in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on:

June 1, 2004
Date

Christopher R. Lewis

LRB_I:\JMYT\234US1\RESPNOTINCOMPAPP.DOC

PTO/SB/05 (05-03) (AW 07-03)
Approved for use through 04/30/2003. OMB 0651-0032
U.S. Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE

UTILITY PATENT APPLICATION TRANSMITTAL

(Only for new nonprovisional applications under 37 C.F.R. 1.53(b))

Attorney Docket No.	JMYT-234US1
First Inventor	Dharshini C. Fongalland
Title	SUBSTRATE
Express Mail Label No.	EV 325926938 US

APPLICATION ELEMENTS

See MPEP chapter 600 concerning utility patent application contents.

1. ☒ Fee Transmittal Form (e.g., PTO/SB/17) *(Submit an original and a duplicate for fee processing*
2. ☐ Applicant claims small entity status. See 37 CFR 1.27.
3. ☒ Specification [*Total Pages* 18]
   (preferred arrangement set forth below
   - Descriptive title of the Invention
   - Cross References to Related Applications
   - Statement Regarding Fed sponsored R & D
   - Reference to sequence listing, a table, or a computer program listing appendix
   - Background of the Invention
   - Brief Summary of the Invention
   - Brief Description of the Drawings *(if filed)*
   - Detailed Description
   - Claim(s)
   - Abstract of the Disclosure
4. ☐ Drawing(s) (*35 U.S.C.113*) [*Total Sheets*]
5. Oath or Declaration [*Total Pages* 3]
   a. ☐ Newly executed (original or copy)
   b. ☒ Copy from a prior application (37 CFR 1.63 (d)) *(for a continuation/divisional with Box 18 completed)*
      i. ☐ DELETION OF INVENTOR(S) Signed statement attached deleting inventor(s) named in the prior application, see 37 CFR 1.63(d)(2) and 1.33(b).
   c. ☐ Unexecuted Declaration
6. ☐ Application Data Sheet. See 37 CFR 1.76

ADDRESS TO: Commissioner for Patents
Mail Stop Patent Application
P.O. Box 1450
Alexandria, VA 22313-1450

7. ☐ CD-ROM or CD-R in duplicate, large table or Computer Program *(Appendix)*
8. Nucleotide and/or Amino Acid Sequence Submission *(if applicable, all necessary)*
   a. ☐ Computer Readable Form (CRF)
   b. Specification Sequence Listing on:
      i. ☐ CD-ROM or CD-R (2 copies); or
      ii. ☐ paper
   c. ☐ Statements verifying identity of above copies

ACCOMPANYING APPLICATIONS PARTS

9. ☐ Assignment Papers (cover sheet & document(s))
10. ☐ 37 C.F.R.§3.73(b) Statement *(when there is an assignee)* ☐ Power of Attorney
11. ☐ English Translation Document *(if applicable)*
12. ☒ Information Disclosure Statement (IDS)/PTO-1449 ☐ Copies of IDS Citations
13. ☒ Preliminary Amendment
14. ☒ Return Receipt Postcard (MPEP 503) *(Should be specifically itemized)*
15. ☐ Certified Copy of Priority Document(s) *(if foreign priority is claimed)*
16. ☐ Nonpublication Request under 35 U.S.C. 122 (b)(2)(B)(i). Applicant must attach form PTO/SB/35 or its equivalent.
17. ☒ Other: 1) Copy of the Preliminary Amendment from the parent application
    2) Certificate of Mailing by Express Mail
    3) Copy of Petition for Extension of Time from parent application

18. If a CONTINUING APPLICATION, *check appropriate box, and supply the requisite information below and in a preliminary amendment, or in an Application Data Sheet under 37 CFR 1.76:*

☒ Continuation ☐ Divisional ☐ Continuation-in-part (CIP) of prior application No: 09 / 807,353

Prior application information *Examiner* Krishnan S. Menon *Group / Art Unit:* 1723

For CONTINUATION or DIVISIONAL APPS only: The entire disclosure of the prior application, from which an oath or declaration is supplied under Box 5b, is considered a part of the disclosure of the accompanying continuation or divisional application and is hereby incorporated by reference. The incorporation can only be relied upon when a portion has been inadvertently omitted from the submitted application parts.

19. CORRESPONDENCE ADDRESS

☒ *Insert Customer Number* **23122** *OR* ☐ *Correspondence address below*

Name					
Address					
City		*State*		*Zip Code*	
Country		*Telephone*		*Fax*	

Name (Print/Type)	Christopher R. Lewis	*Registration No. (Attorney/Agent)*	36,201
Signature		*Date*	October 20, 2003

PTO/SB/17 (05-03) (AW 07/03)
Approved for use through 04/30/2003. OMB 0651-0032
U.S. Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE

FEE TRANSMITTAL for FY 2003

Effective 01/01/2003. Patent fees are subject to annual revision.

☐ Applicant claims small entity status. See 37 CFR 1.27

TOTAL AMOUNT OF PAYMENT ($) 770

Complete If Known	
Application Number	To Be Assigned
Filing Date	Herewith
First Named Inventor	Dharshini C. Fongalland
Examiner Name	
Art Unit	
Attorney Docket No.	JMYT-234US1

METHOD OF PAYMENT *(check all that apply)*

☒ Check ☐ Credit Card ☐ Money Order ☐ Other ☐ None

☒ Deposit Account:

Deposit Account Number: 18-0350

Deposit Account Name: RatnerPrestia

The Commissioner is authorized to: *(check all that apply)*
- ☐ Charge fee(s) indicated below
- ☒ Credit any overpayments
- ☒ Charge any additional fee(s) during the pendency of this application
- ☐ Charge fee(s) indicated below, **except for the filing fee** to the above-identified deposit account.

FEE CALCULATION

1. BASIC FILING FEE

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description	Fee Paid
1001	770	2001	385	Utility filing fee	770
1002	340	2002	170	Design filing fee	
1003	530	2003	265	Plant filing fee	
1004	750	2004	385	Reissue filing fee	
1005	160	2005	80	Provisional filling fee	

SUBTOTAL (1) ($) 770

2. EXTRA CLAIM FEES FOR UTILITY AND REISSUE

			Extra Claims		Fee from below		Fee Paid
Total Claims	18	-20** =	0	X		=	0
Independent Claims	3	-3** =	0	X		=	0
Multiple Dependent				X		=	0

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description
1202	18	2202	9	Claims in excess of 20
1201	86	2201	43	Independent claims in excess of 3
1203	290	2203	145	Multiple dependent claim, if not paid
1204	86	2204	43	** Reissue independent claims over original patent
1205	18	2205	9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) ($) 0

**or number previously paid, if greater; For Reissues, see above

FEE CALCULATION (continued)

3. ADDITIONAL FEES

Large Entity Fee Code	Large Entity Fee ($)	Small Entity Fee Code	Small Entity Fee ($)	Fee Description	Fee Paid
1051	130	2051	65	Surcharge - late filing fee or oath	
1052	50	2052	25	Surcharge - late provisional filing fee or cover sheet.	
1053	130	1053	130	Non-English specification	
1812	2,520	1812	2,520	For filing a request for *ex parte* reexamination	
1804	920*	1804	920*	Requesting publication of SIR prior to Examiner action	
1805	1,840*	1805	1,840*	Requesting publication of SIR after Examiner action	
1251	110	2251	55	Extension for reply within first month	
1252	420	2252	210	Extension for reply within second month	
1253	950	2253	475	Extension for reply within third month	
1254	1,480	2254	740	Extension for reply within fourth month	
1255	2,010	2255	1,005	Extension for reply within fifth month	
1401	330	2401	165	Notice of Appeal	
1402	330	2402	165	Filing a brief in support of an appeal	
1403	290	2403	145	Request for oral hearing	
1451	1,510	1451	1,510	Petition to institute a public use proceeding	
1452	110	2452	55	Petition to revive – unavoidable	
1453	1,330	2453	665	Petition to revive – unintentional	
1501	1,330	2501	665	Utility issue fee (or reissue)	
1502	480	2502	240	Design issue fee	
1503	640	2503	320	Plant issue fee	
1460	130	1460	130	Petitions to the Commissioner	
1807	50	1807	50	Processing fee under 37 CFR 1.17(q)	
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8021	40	8021	40	Recording each patent assignment per property (times number of properties)	
1809	770	2809	385	Filing a submission after final rejection (37 CFR § 1.129(a))	
1810	770	2810	385	For each additional invention to be examined (37 CFR § 1.129(b))	
1801	770	2801	385	Request for Continued Examination (RCE)	
1802	900	1802	900	Request for expedited examination of a design application	
Other fee (specify)					

*Reduced by Basic Filing Fee Paid SUBTOTAL (3) ($) 0

SUBMITTED BY

					Complete (if applicable)
Name (Print/Type)	Christopher R. Lewis	Registration No. Attorney/Agent	36,201	Telephone	610-407-0700
Signature				Date	October 20, 2003

WARNING: Information on this form may become public. Credit card information should not be included on this form. Provide credit card information and authorization on PTO-2038.

This collection of information is required by 37 CFR 1.17 and 1.27. The information is required to obtain or retain a benefit by the public which is to file (and by the USPTO to process) an application. Confidentiality is governed by 35 U.S.C. 122 and 37 CFR 1.14. This collection is estimated to take 12 minutes to complete, including gathering, preparing, and submitting the completed application form to the USPTO. Time will vary depending upon the individual case. Any comments on the amount of time you require to complete this form and/or suggestions for reducing this burden, should be sent to the Chief Information Officer, U.S. Patent and Trademark Office, U.S. Department of Commerce, P.O. Box 1450, Alexandria, VA 22313-1450. DO NOT SEND FEES OR COMPLETED FORMS TO THIS ADDRESS. **SEND TO: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450.**

If you need assistance in completing the form, call 1-800-PTO-9199 (1-800-786-9199) and select option 2.

JMYT-234US

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant:	Dharshini C. Fongalland et al.	: Art Unit:
Application No.:	To Be Assigned	: Examiner:
Filed:	Herewith	:
FOR:	SUBSTRATE	:

PRELIMINARY AMENDMENT

Assistant Commissioner for Patents
Washington, DC 20231

SIR:

Prior to examination, please amend the above-identified application as follows.

IN THE SPECIFICATION:

Please replace the paragraph beginning at page 5, line 8, with the following:

The substrate of the present invention is suitable for use in the preparation of a composite membrane for use in a fuel cell. When for use in a fuel cell, the total thickness of the membrane is suitably less than 200μm and preferably less than 100μm.

Please replace the paragraph beginning at page 6, line 11, with the following:

2) Perfluorinated or partially fluorinated polymers containing aromatic rings such as those described in WO 95/08581 and WO 97/25369 (Ballard Power Systems) which have been functionalised with SO_3H, PO_2H_2, PO_3H_2, $CH_2PO_3H_2$, COOH, OSO_3H, OPO_2H_2, OPO_3H_2. Also included are radiation or chemically grafted perfluorinated polymers, in which a perfluorinated carbon chain, for example, PTFE, fluorinated ethylene-propylene (FEP), tetrafluoroethylene-

ethylene (ETFE) copolymers, tetrafluoroethylene-perfluoroalkoxy (PFA) copolymers, poly (vinyl fluoride) (PVF) and poly (vinylidene fluoride) (PVDF) is activated by radiation or chemical initiation in the presence of a monomer, such as styrene, which can be functionalised to contain an ion exchange group.

IN THE CLAIMS:

Please replace claims 3-15, 17 and 18 with the following amended claims.

3. (Amended) A substrate according to claim 1, wherein the mixed amorphous silica fibres comprise one or more chopped strand(s) of amorphous silica.

4. (Amended) A substrate according to claim 1, wherein the amorphous silica fibres comprise a mixture of both microfibres and chopped fibres in the range of from 95:5% to 5:95% by weight of the mixture respectively.

5. (Amended) A substrate according to claim 4, wherein the amorphous silica fibres comprise a mixture of both microfibres and chopped fibres in the range of from 70:30% to 30:70% by weight of the mixture respectively.

6. (Amended) A substrate according to claim 1, wherein the fibres have a diameter in the range of from 0.1μm to 50μm.

7. (Amended) A substrate according to claim 6, wherein the fibres have a diameter in the range of 0.4μm to 9μm.

8. (Amended) A substrate according to claim 1, wherein the binder comprises a solution or dispersion of ion-exchange polymeric materials, non-ion-conducting polymers, or inorganic materials or mixtures thereof.

9. (Amended) A substrate according to claim 1 for use in the preparation of a composite membrane.

10. (Amended) A composite membrane comprising a porous substrate of fibres and at least one ion-conducting polymer, characterised in that the

substrate comprises a porous matrix of mixed amorphous silica fibres bound with a binder.

11. (Amended) A composite membrane according to claim 10, which when dried then boiled in water undergoes less than or equal to about ±9% change in the area.

12. (Amended) A composite membrane according to claim 10, wherein the total thickness of the membrane is less that 200μm.

13. (Amended) A composite membrane according to claim 10 for use in a fuel cell.

14. (Amended) A process for the manufacture of a substrate, comprising the steps of

(a) dispersing mixed amorphous silica fibres in water to form a slurry;

(b) depositing the slurry onto a mesh bed to form a network;

(c) drying and compacting the fibre network; and

(d) applying, before or after step (c), a dispersion of binder.

15. (Amended) A process for the manufacture of a membrane, comprising the steps of

(i) forming a porous substrate according to claim 14; and thereafter,

(ii) impregnating the porous substrate with a polymeric material to produce a membrane.

17. (Amended) A membrane electrode assembly comprising a composite membrane according to claim 10.

18. (Amended) A fuel cell comprising a composite membrane according to claim 10.

Please add the following new claim:

19. (Newly Added) A process according to claim 15, wherein mixed amorphous silica fibres are randomly oriented in said porous substrate.

Respectfully submitted,

Christopher R. Lewis, Reg. No. 36,201
Attorney for Applicants

CRL/lrb
Dated: April 12, 2001

Suite 301
One Westlakes, Berwyn
P.O. Box 980
Valley Forge, PA 19482-0980
(610) 407-0700

The Assistant Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. 18-0350 of any fees associated with this communication.

EXPRESS MAIL Mailing Label No.: EL751749208US
Date of Deposit: April 12, 2001

I hereby certify that this paper and fee are being deposited, under 37 C.F.R. § 1.10 and with sufficient postage, using the "Express Mail Post Office to Addressee" service of the United States Postal Service on the date indicated above and that the deposit is addressed to the Assistant Commissioner for Patents, Washington, D.C. 20231.

Kathleen Libby

VERSION WITH MARKINGS TO SHOW CHANGES MADE

IN THE SPECIFICATION:

Specification at page 5, line 8:

The substrate of the present invention is [suitably] suitable for use in the preparation of a composite membrane for use in a fuel cell. When for use in a fuel cell, the total thickness of the membrane is suitably less than 200μm and preferably less than 100μm.

Specification at page 6, line 11:

2) Perfluorinated or partially fluorinated polymers containing aromatic rings such as those described in WO 95/08581[, WO 95/08581] and WO 97/25369 (Ballard Power Systems) which have been functionalised with SO_3H, PO_2H_2, PO_3H_2, $CH_2PO_3H_2$, COOH, OSO_3H, OPO_2H_2, OPO_3H_2. Also included are radiation or chemically grafted perfluorinated polymers, in which a perfluorinated carbon chain, for example, PTFE, fluorinated ethylene-propylene (FEP), tetrafluoroethylene-ethylene (ETFE) copolymers, tetrafluoroethylene-perfluoroalkoxy (PFA) copolymers, poly (vinyl fluoride) (PVF) and poly (vinylidene fluoride) (PVDF) is activated by radiation or chemical initiation in the presence of a monomer, such as styrene, which can be functionalised to contain an ion exchange group.

IN THE CLAIMS:

3. (Amended) A substrate according to claim 1 [or claim 2], wherein the mixed amorphous silica fibres comprise one or more chopped strand(s) of amorphous silica.

4. (Amended) A substrate according to [any preceding] claim 1, wherein the amorphous silica fibres comprise a mixture of both microfibres and

chopped fibres in the range of from 95:5% to 5:95% by weight of the mixture respectively.

5. (Amended) A substrate according to claim 4, wherein the amorphous silica fibres comprise a mixture of both microfibres and chopped fibres in the range of from 70:30% to 30:70% by weight of the mixture respectively.

6. (Amended) A substrate according to [any preceding] claim 1, wherein the fibres have a diameter in the range of from 0.1μm to 50μm.

7. (Amended) A substrate according to claim 6, wherein the fibres have a diameter in the range of 0.4μm to 9μm.

8. (Amended) A substrate according to [any preceding] claim 1, wherein the binder comprises a solution or dispersion of ion-exchange polymeric materials, non-ion-conducting polymers, or inorganic materials or mixtures thereof.

9. (Amended) A substrate according to [any preceding] claim 1 for use in the preparation of a composite membrane.

10. (Amended) A composite membrane comprising a porous substrate of fibres and at least one ion-conducting polymer, characterised in that the substrate [is one according to any preceding claim, which] comprises a porous matrix of mixed amorphous silica fibres bound with a binder.

11. (Amended) A composite membrane according to claim 10, which when [tested by the method described herein in the Examples, results in] dried then boiled in water undergoes less than or equal to about ±9% change in the area.

12. (Amended) A composite membrane according to claim 10, [or claim 11] wherein the total thickness of the membrane is less that 200μm.

13. (Amended) A composite membrane according to [any one of claims] claim 10 [to 12] for use in a fuel cell.

14. (Amended) A process for the manufacture of a substrate [according to any one of claims 1 to 9], [which process comprises] comprising the steps of

(a) dispersing [the] mixed amorphous silica fibres in water to form a slurry;

(b) depositing the slurry onto a mesh bed to form a network;

(c) drying and compacting the fibre network; and

(d) applying, before or after step (c), a dispersion of binder.

15. (Amended) A process for the manufacture of a membrane [according to any one of claims 10 to 13], [which process comprises] comprising the steps of

(i) forming a porous substrate [of, preferably randomly orientated individual mixed amorphous silica fibres bound with a binder by a process] according to claim 14; and thereafter,

(ii) impregnating the porous substrate with a polymeric material to produce a membrane.

17. (Amended) A membrane electrode assembly comprising [a substrate according to any one of claim 1 to 9 and/or] a composite membrane according to [any one of claims] claim 10 [to 13].

18. (Amended) A fuel cell comprising [a substrate according to any one of claim 1 to 9 and/or] a composite membrane according to [any one of claims] claim 10 [to 13].

Claim 19 has been added.

Declaration and Power of Attorney For Patent Application
English Language Declaration

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name,

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled
SUBSTRATE,
the specification of which is attached hereto unless the following box is checked:

☒ was filed on September 3, 1999 as
United States Application Number or PCT International Application Number PCT/GB99/02935
and was amended by Preliminary Amendment filed along with the application (if applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56.

I hereby claim foreign priority benefits under 35 U.S.C. §119(a)-(d) or § 365(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed:

Prior Foreign Application(s)			Priority Not Claimed
9822569.1	Great Britain	16 October 1998	☐
(Number)	(Country)	(Day/Month/Year Filed)	
_____	_____	_____	☐
(Number)	(Country)	(Day/Month/Year Filed)	

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below.

_____	_____
(Application Number)	(Filing Date)
_____	_____
(Application Number)	(Filing Date)

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112, I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT international filing date of this application:

(Application Number)	(Filing Date)	(Status - patented, pending, abandoned)
(Application Number)	(Filing Date)	(Status - patented, pending, abandoned)

POWER OF ATTORNEY: As a named inventor, I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith:

Paul F. Prestia	Reg. No. 23,031	Lawrence E. Ashery	Reg. No. 34,515	Jack J. Jankovitz	Reg. No. 42,690
Allan Ratner	Reg. No. 19,717	Christopher R. Lewis	Reg. No. 36,201	Jonathan H. Spadt	Reg. No. 45,122
Andrew L. Ney	Reg. No. 20,300	Robert L. Andersen	Reg. No. 25,771	Christopher I. Halliday	Reg. No. 42,621
Kenneth N. Nigon	Reg. No. 31,549	Joshua L. Cohen	Reg. No. 38,040	Scott A. Mckeown	Reg. No. 42,866
Kevin R. Casey	Reg. No. 32,117	Daniel N. Calder	Reg. No. 27,424	Stanley N. Protigal	Reg. No. 28,657
Benjamin E. Leace	Reg. No. 33,412	Louis W. Beardell, Jr.	Reg. No. 40,506		
James C. Simmons	Reg. No. 24,842	Jacques L. Etkowicz	Reg. No. 41,738		

Address all correspondence to: Christopher R. Lewis
Ratner & Prestia, Suite 301, One Westlakes, Berwyn, P.O. Box 980, Valley Forge, PA 19482-0980
Address all telephone calls to: Christopher R. Lewis at (610) 407-0700.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Full name of sole or first inventor (given name, family name) Dharshini Chryshantha FONGALLAND

Inventor's signature _____ Date 23/03/01

Residence Slough, United Kingdom

Citizenship Sri Lankan

Post Office Address 174 Windsor Road
Slough SL1 2JD, United Kingdom

Full name of second joint inventor, if any (given name, family name) John Malcolm GASCOYNE

Second Inventor's signature _____ Date 28th March 01

Residence Bucks, United Kingdom

Citizenship British

Post Office Address Lyduska, Routs Green, Bledlow Ridge, High Wycombe
Bucks HP14 4BB, United Kingdom

☒ Additional inventors are being named on separately numbered sheets attached hereto.

Full name of third joint inventor, if any (given name, family name) Thomas Robertson RALPH

Third inventor's signature Thomas Ralph Date 20/3/01

Residence Reading, United Kingdom

Citizenship British

Post Office Address 94 Shaftesbury Road

Reading RG30 2QJ, United Kingdom

Full name of fourth joint inventor, if any (given name, family name) _____

Fourth inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

_____

Full name of fifth joint inventor, if any (given name, family name) _____

Fifth inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

_____

Full name of sixth joint inventor, if any (given name, family name) _____

Sixth inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

_____

Full name of seventh joint inventor, if any (given name, family name) _____

Seventh inventor's signature _____ Date _____

Residence _____

Citizenship _____

Post Office Address _____

_____

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appln. No:	To Be Assigned
Applicant:	Dharshini C. Fongalland et al.
Filed:	Herewith
Title:	SUBSTRATE
TC/A.U.:	
Examiner:	
Confirmation No.:	
Docket No.:	JMYT-234US1

Continuation of:

Appln. No:	09/807,353
Applicant:	Dharshini C. Fongalland et al.
Filed:	April 12, 2001
Title:	SUBSTRATE
TC/A.U.:	1723
Examiner:	Krishnan S. Menon
Confirmation No.:	6310
Docket No.:	JMYT-234US

SUPPLEMENTAL PRELIMINARY AMENDMENT

Mail Stop Patent Application
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

Prior to examination, please amend the above-identified application as follows:

☒ **Amendments to the Specification** begin on page 2 of this paper.

☒ **Amendments to the Claims** are reflected in the listing of claims which begins on page 3 of this paper.

☐ **Amendments to the Drawings** begin on page of this paper and include an attached replacement sheet(s).

☐ **Amendments to the Abstract** are on page of this paper. A clean version of the Abstract is on page of this paper.

☒ **Remarks/Arguments** begin on page 6 of this paper.

Amendments to the Specification:

Please add the following new paragraph at page 1, after the title of the application:

This application is a continuation of U.S. Patent Application No. 09/807,353, filed April 12, 2001, which is incorporated by reference herein.

Amendments to the Claims: This listing of claims will replace all prior versions, and listings, of claims in the application

Listing of Claims:

1. (Currently Amended) A substrate, suitable for the preparation of a composite membrane, which substrate comprises a porous ~~matrix~~ non-woven sheet of fibres, ~~characterised in that~~ wherein the fibres comprise ~~mixed amorphous silica fibres~~ a mixture of micro-fine amorphous silica fibres and one or more chopped strand(s) of amorphous silica ~~that are~~ and the fibres are bound with a binder.

2. (Canceled)

3. (Canceled)

4. (Currently Amended) A substrate according to claim 1, wherein the ~~amorphous silica fibres comprise a~~ mixture ~~of both~~ comprises microfibres and chopped fibres in the range of from 95:5% to 5:95% by weight of the mixture respectively.

5. (Currently Amended) A substrate according to claim 4, wherein the ~~amorphous silica fibres comprise a~~ mixture ~~of both~~ comprises microfibres and chopped fibres in the range of from 70:30% to 30:70% by weight of the mixture respectively.

6. (Original) A substrate according to claim 1, wherein the fibres have a diameter in the range of from 0.1μm to 50μm.

7. (Original) A substrate according to claim 6, wherein the fibres have a diameter in the range of 0.4μm to 9μm.

8. (Original) A substrate according to claim 1, wherein the binder comprises a solution or dispersion of ion-exchange polymeric materials, non-ion-conducting polymers, or inorganic materials or mixtures thereof.

9. (Original) A substrate according to claim 1 for use in the preparation of a composite membrane.

10. (Currently Amended) A composite membrane comprising a porous substrate of fibres and at least one ion-conducting polymer, ~~characterised in that~~ wherein the substrate comprises

a porous ~~matrix~~ non-woven sheet of ~~mixed amorphous silica fibres~~ a mixture of micro-fine amorphous silica fibres and one or more chopped strand(s) of amorphous silica and the fibres are bound with a binder.

11. (Original) A composite membrane according to claim 10, which when dried then boiled in water undergoes less than or equal to about ±9% change in the area.

12. (Original) A composite membrane according to claim 10, wherein the total thickness of the membrane is less that 200μm.

13. (Original) A composite membrane according to claim 10 for use in a fuel cell.

14. (Currently Amended) A process for the manufacture of a substrate, comprising the steps of

(a) dispersing ~~mixed amorphous silica fibres~~ a mixture of micro-fine amorphous silica fibres and one or more chopped strand(s) of amorphous silica in water to form a slurry;

(b) depositing the slurry onto a mesh bed to form a fibre network;

(c) drying and compacting the fibre network; and

(d) applying, before or after step (c), a dispersion of binder.

15. (Original) A process for the manufacture of a membrane, comprising the steps of

(i) forming a porous substrate according to claim 14; and thereafter,

(ii) impregnating the porous substrate with a polymeric material to produce a membrane.

16. (Original) A process according to claim 15, wherein step (ii) is carried out by nip roller coating of the substrate to fill it with a solution of ion-conducting polymeric material, and further compaction and drying of the membrane.

17. (Original) A membrane electrode assembly comprising a composite membrane according to claim 10.

18. (Original) A fuel cell comprising a composite membrane according to claim 10.

19. (Currently Amended) A process according to claim 15, wherein ~~mixed amorphous silica~~ the fibres are randomly oriented in said porous substrate.

20. (New) A substrate according to claim 1, wherein the fibres are randomly oriented.

Remarks/Arguments:

Claims 1 and 4-20 are the pending claims in this application. With this filing, the applicants respectfully submit that any rejection under 35 U.S.C. § 103 using Denton should be overcome based on Section 103(c).

Respectfully submitted,

Christopher R. Lewis, Reg. No. 36,201
Attorney for Applicants

CRL/lrb

Dated: October 20, 2003

P.O. Box 980
Valley Forge, PA 19482
(610) 407-0700

The Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. **18-0350** of any fees associated with this communication.

EXPRESS MAIL Mailing Label No.: EV 325926938 US
Date of Deposit: October 20, 2003

I hereby certify that this correspondence is being deposited with the United States Postal Service as first class mail, with sufficient postage, in an envelope addressed to: Mail Stop Patent Application, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on:

Kathleen Libby

JMYT-234US1

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appln. No:	To Be Assigned
Applicant:	Dharshini C. Fongalland et al.
Filed:	Herewith
Title:	SUBSTRATE
TC/A.U.:	
Examiner:	
Confirmation No.:	
Docket No.:	JMYT-234US1

Continuation of:

Appln. No:	09/807,353
Applicant:	Dharshini C. Fongalland et al.
Filed:	April 12, 2001
Title:	SUBSTRATE
TC/A.U.:	1723
Examiner:	Krishnan S. Menon
Confirmation No.:	6310
Docket No.:	JMYT-234US

INFORMATION DISCLOSURE STATEMENT

Mail Stop Patent Application
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

Pursuant to 37 C.F.R. §§ 1.97 and 1.98 and to the duty of disclosure set forth in 37 C.F.R. § 1.56, the Examiner in charge of the above-identified application is requested to consider and make of record the references listed on the PTO 1449 (RP) submitted herewith.

Although the information submitted herewith may be "material" to the Examiner's consideration of the subject application, this submission is not intended to constitute an admission that such information is "prior art" as to the claimed invention.

In accordance with 37 C.F.R. § 1.97(g), the filing of this Information Disclosure Statement shall not be construed to mean that a search has been made.

Under 37 C.F.R. § 1.98(d), copies of the patents and publications listed on the enclosed PTO Form 1449 are not required to be provided, because they were cited by or submitted to the Patent and Trademark Office in prior application Serial No. 09/807,353, filed April 12, 2001, which is relied upon for an earlier filing date under 35 U.S.C. § 120.

This Information Disclosure Statement is being filed concurrently with the above-referenced continuation application.

Respectfully submitted,

Christopher R. Lewis, Reg. No. 36,201
Attorney for Applicants

CRL/lrb
Enclosure: Form PTO/SB/08 (3 pgs.)

Dated: October 20, 2003

P.O. Box 980
Valley Forge, PA 19482
(610) 407-0700

The Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. **18-0350** of any fees associated with this communication.

EXPRESS MAIL Mailing Label No.: EV 325926938 US
Date of Deposit: October 20, 2003

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Kathleen Libby

LRB_I:\JMYT\234US1\IDS-001.DOC

(AW 08/03)

Substitute for Form 1449A/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

Complete if Known	
Application Number	To Be Assigned
Filing Date	Herewith
First Named Inventor	Dharshini C. Fongalland
Art Unit	
Examiner Name	
Attorney Docket No.	JMYT-234US1

SHEET 1 of 3

U.S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number: Number - Kind Code[2] (if known)	Publication Date (MM-DD-YYYY)	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-3,282,875	11-01-1966	Connolly et al.	
		US-4,329,435	05-11-1982	Kimoto et al.	
		US-4,330,654	05-18-1982	Ezzell et al.	
		US-4,358,545	11-09-1982	Ezzell et al.	
		US-4,417,969	11-29-1983	Ezzell et al.	
		US-4,433,082	02-21-1984	Grot	
		US-4,610,762	09-09-1986	Birdwell	
		US-4,842,620	06-27-1989	Hammel et al.	
		US-4,940,525	07-10-1990	Ezzell et al.	
		US-5,094,995	03-10-1992	Butt et al.	
		US-5,438,082	08-01-1995	Helmer-Metzmann et al.	
		US-5,468,574	11-21-1995	Ehrenberg et al.	
		US-5,523,181	06-04-1996	Stonehart et al.	
		US-5,547,551	08-20-1996	Bahar et al.	
		US-5,595,676	01-21-1997	Barnes et al.	
		US-5,599,639	02-04-1997	Sansone et al.	
		US-6,042,958	03-28-2000	Denton et al.	

FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document: Country Code[3] - Number[4] - Kind Code[5] (If known)	Publication Date (MM-DD-YYYY)	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		EP-0 331 321	09-06-1989	ICI PLC		☐
		EP-0 345 964	12-13-1989	ICI PLC		☐
		EP-0 574 791	12-22-1993	Hoechst AG		☐
		EP-0 731 520	09-11-1996	Johnson Matthey PLC		☐
		EP-0 791 974	08-27-1997	Johnson Matthey PLC		☐
		EP-0 875 524	11-04-1998	DSM NV		☐

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to Applicant.

[1]Applicant's unique citation designation number (optional).

[2]See Kind Codes of USPTO Patent Documents at www.uspto.gov or MPEP 901.04.

[3]Enter Office that issued the document, by the two-letter code (WIPO Standard St.3).

[4]For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document.

[5]Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible.

[6]Applicant is to place a check mark here if English language translation is attached.

PTO/SB/08a (05-03) (AW 07/03)
Approved for use through 05/31/03. OMB 0651-0031
U.S. Patent and Trademark Office: U.S. DEPARTMENT OF COMMERCE

Substitute for Form 1449A/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

SHEET 2 of 3

Complete if Known	
Application Number	To Be Assigned
Filing Date	Herewith
First Named Inventor	Dharshini C. Fongalland
Art Unit	
Examiner Name	
Attorney Docket No.	JMYT-234US1

U.S. PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Document Number: Number - Kind Code[2] (if known)	Publication Date (MM-DD-YYYY)	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear
		US-			
		US-			
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FOREIGN PATENT DOCUMENTS

Examiner Initials*	Cite No.[1]	Foreign Patent Document: Country Code[3] - Number[4] - Kind Code[5] (If known)	Publication Date (MM-DD-YYYY)	Name of Patentee or Applicant of Cited Document	Pages, Columns, Lines, Where Relevant Passages or Relevant Figures Appear	T[6]
		DD-283 478	10-17-1990	Leipzig Chemieanlagen		☐
		GB-1 599 077	09-30-1981	Yuasa Battery Co. Ltd.		☐
		JP-57 159502 (Abstract Only)	10-01-1982	Toyo Boseki KK		☐
		JP-06 304548 (Abstract Only)	11-01-1994	Matsushita Electric Ind. Co. Ltd.		☐
		NL-8003824	02-01-1982	Akzo NV		☐
		WO-94/16002	07-21-1994	Allied Signal, Inc.		☐
		WO-95/08581	03-30-1995	Ballard Power Systems		☐
		WO-97/25369	07-17-1997	Ballard Power Systems, C. Stone, A. E. Steck		☐

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to Applicant.

[1]Applicant's unique citation designation number (optional).

[2]See Kind Codes of USPTO Patent Documents at www.uspto.gov or MPEP 901.04.

[3]Enter Office that issued the document, by the two-letter code (WIPO Standard St.3).

[4]For Japanese patent documents, the indication of the year of the reign of the Emperor must precede the serial number of the patent document.

[5]Kind of document by the appropriate symbols as indicated on the document under WIPO Standard ST.16 if possible.

[6]Applicant is to place a check mark here if English language translation is attached.

(AW 08/03)

Substitute for Form 1449B/PTO

INFORMATION DISCLOSURE STATEMENT BY APPLICANT

(Use as many sheets as necessary)

SHEET 3 of 3

Complete if Known	
Application Number	To Be Assigned
Filing Date	Herewith
First Named Inventor	Dharshini C. Fongalland
Art Unit	
Examiner Name	
Attorney Docket No.	JMYT-234US1

NON-PATENT LITERATURE DOCUMENTS

Examiner Initials*	Cite No.[1]	Include name of the author (in CAPITAL LETTERS), title of the article (when appropriate), title of the item (book, magazine, journal, serial, symposium, catalog, etc.), date, page(s), volume-issue number(s), publisher, city and/or country where published	T[2]
		Kolde et al., "Advanced Composite Polymer Electrolyte Fuel Cell Membranes," Electrochemical Society Proceedings, Vol. 95-23 (1995) pp. 193-201.	☐
		International Search Report dated January 12, 2000, from International Application No. PCT/GB99/02935	☐
		British Search Report dated February 19, 1999, from British Application No. 9822569.1.	☐
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			☐
			☐

Examiner Signature		Date Considered	

*EXAMINER: Initial if reference considered, whether or not citation is in conformance with MPEP 609. Draw line through citation if not in conformance and not considered. Include copy of this form with next communication to Applicant.

[1]Applicant's unique citation designation number (optional).

[2]Applicant is to place a check mark here if English language translation is attached.

PTO/SB/22 (05-03) (AW 07/03)
Approved for use through 10/31/2002. OMB 0651-0031
U.S. Patent and Trademark Office; U.S. DEPARTMENT OF COMMERCE
Under the Paperwork Reduction Act of 1995, no persons are required to respond to a collection of information unless it displays a valid OMB control number.

PETITION FOR EXTENSION OF TIME UNDER 37 CFR 1.136(a)	Docket Number (Optional) JMYT-234US

In re Application of Dharshini C. Fongalland et al.	
Application Number 09/807,353	Filed April 12, 2001
For SUBSTRATE	
Art Unit 1723	Examiner Krishnan S. Menon

This is a request under the provisions of 37 CFR 1.136(a) to extend the period for filing a reply in the above identified application.

The requested extension and appropriate non-small-entity fee are as follows (check time period desired):

☐ One month	(37 CFR 1.17(a)(1))	$
☒ Two months	(37 CFR 1.17(a)(2))	$420
☐ Three months	(37 CFR 1.17(a)(3))	$
☐ Four months	(37 CFR 1.17(a)(4))	$
☐ Five months	(37 CFR 1.17(a)(5))	$

☐ Applicant claims small entity status. See 37 CFR 1.27. Therefore, the fee amount shown above is reduced by one-half, and the resulting fee is: $ ___ .

☒ A check in the amount of the fee is enclosed.

☐ Payment by credit card. Form PTO-2038 is attached.

☐ The Director has already been authorized to charge fees in this application to a Deposit Account.

☒ The Director is hereby authorized to charge any fees which may be required or credit any overpayment to Deposit Account Number 18-0350. I have enclosed a duplicate copy of this sheet.

I am the
☐ applicant/inventor.
☐ assignee of record of the entire interest. See 37 CFR 3.71. Statement under 37 CFR 3.73(b) is enclosed (Form PTO/SB/96).
☒ attorney or agent of record.
☐ attorney or agent under 37 CFR 1.34(a). Registration number if acting under 37 CFR 1.34(a) .

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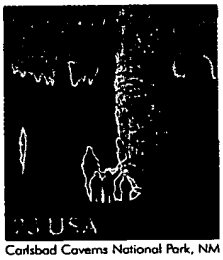
DOCUMENTS UTILITY PATENT APPLICATION TRANSMITTAL (1PG.); FEE TRANSMITTAL (1PG.) IN DUPL.; COPY OF INT'L APP. NO. PCT/GB99/02935 (THE SPECIFICATION (18 PGS.) OF THE PARENT APP.); COPY OF THE PRELIM. AMEND. FROM THE PARENT APP. (7 PGS.); COPY OF THE DECLARATION/POWER OF ATTORNEY FROM THE PARENT APP. (3 PGS.); SUPP. PRELIM. AMEND. (6 PGS.); IDS (2 PGS.) WITH FORM PTO/SB/08 (3 PGS.); CERTIF. OF MAILING BY EXP. MAIL (1 PG.); ADD'L CHECK; ADD'L COPY OF EXTENSION FROM PARENT



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(57) Abstract

A substrate, suitable for the preparation of a composite membrane, which substrate comprises a porous matrix of fibres, characterised in that the fibres comprise mixed amorphous silica fibres that are bound with a binder; a composite membrane comprising the substrate and a process for the preparation of the substrate and composite membrane is disclosed.

SUBSTRATE

The present invention relates to a substrate for a composite membrane that is of use in electrochemical devices, particularly fuel cells, and a process for the manufacture of the substrate and composite membrane.

Electrochemical cells invariably comprise an ion-conducting electrolyte and two electrodes, the anode and cathode, at which the desired electrochemical reactions take place. Electrochemical cells may be found in a range of devices, for example fuel cells, batteries, sensors, electrodialysis reactors and electrolytic reactors. They have a diverse range of applications, including the electrolysis of water, chemical synthesis, salt splitting, water purification, effluent treatment and metal finishing, among others.

A fuel cell is an energy conversion device that efficiently converts the stored chemical energy of its fuel into electrical energy. It does so by combining either hydrogen, stored as a gas or methanol, stored as a liquid or a gas, with oxygen to generate electrical power. The hydrogen or methanol is oxidised at the anode and oxygen is reduced at the cathode. Both electrodes are of the gas diffusion type. The electrolyte has to be in contact with both electrodes, and may be acidic or alkaline, and liquid or solid, in nature. In proton exchange membrane fuel cells (PEMFC), the electrolyte is a solid, ion-conducting, *i.e.* a proton-conducting, polymer membrane. The membrane is commonly based on a copolymer of perfluorosulphonic acid and tetrafluoroethylene. The combined structure formed from the membrane and the two gas diffusion electrodes is known as the membrane electrode assembly (MEA).

Conventionally, solid ion-conducting membrane electrolytes useful in fuel cells and other devices are selected from commercially-available membranes, for example perfluorinated membranes sold under the trade names Nafion® (E I DuPont de Nemours and Co.), Aciplex® (Asahi Chemical Industry) and Flemion® (Asahi Glass KK). For application in the PEMFC, they are typically below 200μm in thickness to ensure a high level of ionic conductivity. One of the problems experienced with these conventional proton-conducting membranes used for PEM fuel cell construction, is the dimensional changes that occur as the

level of water content (hydration) of the membrane changes. This is a particular problem during fabrication of the MEA, in which the membrane is typically in a highly hydrated form, as the stresses produced by changes in hydration during the conventionally-employed thermal bonding process can be large enough to break the bond between either the catalyst and the membrane or the catalyst and the substrate. Furthermore, these dimensional changes lead to considerable difficulties in handling membranes during the fabrication of MEAs, particularly large area MEAs in excess of, for example, 500cm^2. The thinner the membrane, the more difficult the handling becomes.

Yet further, it is current practice that most MEAs are fabricated as single items, with areas of, for example, 500cm^2 in a batch-type process. It is critical to the successful commercialisation of the PEMFC that lower cost, high volume, MEA manufacturing processes be developed in the future, such as a continuous fabrication process. The problem of dimensional change of the membrane with changes in hydration on a continuous process, which may employ membranes of many hundreds of metres in length, would then be an even more serious issue, and would add significant complications and cost to the manufacturing process.

With thicker types of membrane (*e.g.* >350μm) developed for other applications, it has been possible to incorporate 'macro' reinforcing materials, such as woven polytetrafluoroethylene (PTFE), to minimise such dimensional changes. However, these thicker materials have too low an ionic conductivity to be of use in the PEMFC. US patent 5,547,551 describes the fabrication of ultra-thin reinforced membranes, below 25μm in thickness, comprising proton-exchange polymeric material incorporated into an expanded porous PTFE membrane. According to Kolde *et al*, Electrochemical Society Proceedings 95 (23) 193-201 (1995), these reinforced membranes have considerably improved dimensional stability compared to the conventional non-reinforced membranes, such as Nafion® 117 which shows shrinkage upon dehydration from the hydrated state. However, such materials have a higher specific resistance (*i.e.* lower ionic conductivity) by a factor of at least two than a non-reinforced pure proton-conducting membrane such as Nafion® 117.

The higher specific resistance of the above reinforced membranes means that, in practice, they must be much thinner than the equivalent pure proton-conducting membrane to maintain the same overall conductivity and thus cell performance. However, reducing the thickness of the membrane reduces the advantages that a reinforced membrane can provide. For example, there is a limit to the extent to which the thickness of the membrane can be reduced, since the durability and longevity can also decrease, and reactant gas cross-over through the membrane is more liable to occur, leading to a reduction in cell performance. Furthermore, the problems associated with dimensional stability and handling for MEA fabrication can be exacerbated with thinner membranes.

There is therefore the need to overcome the disadvantages of conventional pure and prior art reinforced membranes, by providing a novel composite ion-exchange membrane having a significantly improved dimensional stability and satisfactory handling without compromising the ionic conductivity and reactant gas cross-over parameters. Furthermore, there is a need to take account of the likely process(es) by which the membrane would be manufactured in the future in choosing an appropriate membrane composition. In particular, with the prospect of continuous fabrication processes mentioned above, it is not only the structure of the membrane that may be critical. In a composite membrane generally comprising a porous substrate of fibres impregnated, coated or otherwise associated with the ion-conducting polymer (*e.g.* Nafion®), the strength and stability of the substrate itself would be an important factor.

Accordingly, the present invention provides a substrate, suitable for the preparation of a composite membrane, which substrate comprises a porous matrix of fibres, characterised in that the fibres comprise mixed amorphous silica fibres that are bound with a binder.

The amorphous silica for use in the substrate according to the invention is to be distinguished from crystalline quartz, although there is a tendency in an industrial context for the terms "quartz" and "silica" to be used interchangeably. Although both are chemically silicon dioxide, quartz is the crystalline form and is both hard and brittle, whereas the fibrous materials (the amorphous silica for use in the substrate of the invention) are made from either

natural or synthetic quartz, and are amorphous and glass like in character, having no crystalline structure.

By "mixed amorphous silica fibres" is meant a mixture of both one or more micro-fine amorphous silica fibres and one or more of chopped strands of amorphous silica. For example, chopped silica fibres are available from Quartz et Silice BP, France under the trade name Quartzel. The base filament is available as a continuous fibre in 14μm, 9μm or 7μm diameters and can be supplied as chopped strands in a range of lengths such as 20mm chopped silica fibres. Silica microfibres are available from Johns Manville Insulation Group, Denver, USA, under the trade name of Q-Fibre, such as Q-Fibre Type 106. These are available in a range of nominal fibre diameters from 0.4μm to 4μm. The amount of microfibre and chopped fibres in the mixture is in the range of from 95 to 5% and 5 to 95% by weight of the mixture, respectively. Preferably, the amounts are 90 to 10% and 10 to 90% w/w, respectively. More preferably, they are present in a range 70 to 30% to 30 to 70% w/w, respectively.

The mixed amorphous silica fibres within the substrate are preferably randomly orientated in the x and y direction (in-plane), producing a two-dimensional isotropic structure. Additionally, random orientation in the z direction (through-plane) can be introduced with the inclusion of very short fibres, typically lengths of less than or equal to 0.2mm or very fine fibres, typically of diameters less than or equal to 1μm. The fibres typically have a diameter in the range of from 0.1μm to 50μm, preferably 0.2μm to 20μm and, more preferably, about 0.4μm to 9μm. The fibres typically have lengths in the range of from 0.05mm to 300mm, suitably 0.5mm to 150mm, preferably 1mm to 50mm and, more preferably, about 6mm to 20mm.

The porous substrate typically has at least 50%, suitably at least 75%, of the individual pore sizes being greater than 1μm in at least one direction, although a porous substrate wherein some of the pores are less than 1μm in all directions is within the scope of the invention.

It is also necessary to coat the fibres with one or more different materials after forming the porous substrate network to act as a binder and provide the necessary physical integrity of the structure. Fibres may be coated with a solution or dispersion of ion-exchange polymeric

materials, such as Nafion® 1100EW solution, or other non-ion-conducting polymers such as PTFE, FEP, PVDF, Viton®, polyethylene and polypropylene, such as are further described below, or inorganic materials such as amorphous silica, titania, zirconia, zirconium silicate, zirconium phosphates or the like, or mixtures thereof. Solutions of ion-exchange polymers may be either organic or aqueous, and the polymer may be either in protonic form or in ion-exchanged form, wherein the proton site is replaced with, *e.g.*, Na^+ or t-butylammonium ion.

The substrate of the present invention is suitably for use in the preparation of a composite membrane for use in a fuel cell. When for use in a fuel cell, the total thickness of the membrane is suitably less than 200μm and preferably less than 100μm.

For its use in the preparation of a composite membrane, the substrate is preferably associated with an ion-conducting polymer. Accordingly, the present invention further provides a composite membrane comprising a porous substrate of fibres and at least one ion-conducting polymer, characterised in that the substrate comprises mixed amorphous silica fibres, as defined hereinabove, that are bound with a binder.

The substrates according to the present invention, when used as a membrane by the incorporation of an ion-conducting polymer therein, produce a surprising effect on the dimensional stability of the membrane when subject to full hydration conditions. Accordingly, when tested by the method described hereinafter in the Examples, the dimensional changes in membranes based on the substrates according to the present invention result in less than or equal to about ±9% change in their areas.

For PEM fuel cell applications, the ion-conducting polymer is a proton-conducting polymer, examples of such polymers being well known to those skilled in the art. More than one proton-conducting polymer may be present and/or a non-ion-conducting polymer may also be included in the novel membrane of the invention.

The proton conducting polymers suitable for use in the present invention may include, but are not limited to:

1) Polymers which have structures with a substantially fluorinated carbon chain optionally having attached to it side chains that are substantially fluorinated. These polymers contain sulphonic acid groups or derivatives of sulphonic acid groups, carboxylic acid groups or derivatives of carboxylic acid groups, phosphonic acid groups or derivatives of phosphonic acid groups, phosphoric acid groups or derivatives of phosphoric acid groups and/or mixtures of these groups. Perfluorinated polymers include Nafion®, Flemion® and Aciplex® commercially available from E. I. DuPont de Nemours (U.S. Patents 3,282,875; 4,329,435; 4,330,654; 4,358,545; 4,417,969; 4,610,762; 4,433,082 and 5,094,995), Asahi Glass KK and Asahi Chemical Industry respectively. Other polymers include those covered in U.S. Patent 5,595,676 (Imperial Chemical Industries plc) and U.S. Patent 4,940,525 (Dow Chemical Co.)

2) Perfluorinated or partially fluorinated polymers containing aromatic rings such as those described in WO 95/08581, WO 95/08581 and WO 97/25369 (Ballard Power Systems) which have been functionalised with SO_3H, PO_2H_2, PO_3H_2, $CH_2PO_3H_2$, COOH, OSO_3H, OPO_2H_2, OPO_3H_2. Also included are radiation or chemically grafted perfluorinated polymers, in which a perfluorinated carbon chain, for example, PTFE, fluorinated ethylene-propylene (FEP), tetrafluoroethylene-ethylene (ETFE) copolymers, tetrafluoroethylene-perfluoroalkoxy (PFA) copolymers, poly (vinyl fluoride) (PVF) and poly (vinylidene fluoride) (PVDF) is activated by radiation or chemical initiation in the presence of a monomer, such as styrene, which can be functionalised to contain an ion exchange group.

3) Fluorinated polymers such as those disclosed in EP 0 331 321 and EP 0345 964 (Imperial Chemical Industries plc) containing a polymeric chain with pendant saturated cyclic groups and at least one ion exchange group which is linked to the polymeric chain through the cyclic group.

4) Aromatic polymers such as those disclosed in EP 0 574 791 and US Patent 5,438,082 (Hoechst AG) for example sulphonated polyaryletherketone. Also aromatic polymers such as polyether sulphones which can be chemically grafted with a polymer with ion exchange functionality such as those disclosed in WO 94/16002 (Allied Signal Inc.).

5) Nonfluorinated polymers include those disclosed in U.S. Patent 5,468,574 (Dais Corporation) for example hydrocarbons such as styrene-(ethylene-butylene)- styrene, styrene-(ethylene-propylene)-styrene and acrylonitrile-butadiene-styrene co- and terpolymers where the styrene components are functionalised with sulphonate, phosphoric and/or phosphonic groups.

6) Nitrogen containing polymers including those disclosed in U.S. Patent 5,599,639 (Hoechst Celanese Corporation), for example, polybenzimidazole alkyl sulphonic acid and polybenzimidazole alkyl or aryl phosphonate.

7) Any of the above polymers which have the ion exchange group replaced with a sulphonyl chloride (SO_2Cl) or sulphonyl fluoride (SO_2F) group rendering the polymers melt processable. The sulphonyl fluoride polymers may form part of the precursors to the ion exchange membrane or may be arrived at by subsequent modification of the ion exchange membrane. The sulphonyl halide moieties can be converted to a sulphonic acid using conventional techniques such as, for example, hydrolysis.

Non-ion conducting polymeric materials which may be used in addition to the one or more ion conducting or proton conducting polymers include PTFE, FEP, PVDF, Viton® and hydrocarbon types such as polyethylene, polypropylene and polymethylmethacralate.

Other ion-conducting polymeric materials which are not proton conducting polymers may be used in the filler material. For example, such polymers can be used for applications requiring a bipolar membrane or a completely anion exchange membrane. Anion exchange polymers are generally based on quaternary ammonium groups, rather than the fixed sulphonic acid groups in proton conducting polymers. These include, for example, the tetraalkyl ammonium group ($-N^+R_3$) and the quaternary ammonium centre in Tosflex® membranes ($-N(R_1)(CH_2)_yN^+(R_3)$) supplied by Tosoh. However, it can be envisaged that all of the proton exchange polymers described above could have anion exchange equivalents.

The polymer is suitably applied to the coated fibres (substrate) in the form of a solution, the solvents of which may be either organic or aqueous based. Solvents of all of the above polymers may include or may be modified to include, water, methanol and/or other aliphatic alcohols, ethers, acetone, tetrahydrofuran (THF), n-methyl-pyrrolidone (NMP), dimethyl sulphoxide (DMSO), dimethyl formamide (DMF), dimethyl acetamide (DMAc), or protonic solvents such as sulphuric acid or phosphoric acid, and/or mixtures of the above. However, it has been found that an essentially aqueous solution of the polymer as described in EP 0 731 520 is preferred.

A flexible free-standing, dimensionally stable composite membrane is produced by using the substrate of the present invention, resulting in greater handlability. The membrane of the invention is therefore also more amenable to high volume, continuous production processes, as described hereinafter. The high dimensional stability of the membrane enables thinner membranes to be produced, which are more amenable to higher volume MEA manufacturing processes than are current membranes, at similar thicknesses. Current materials show very large dimensional changes with changes in the levels of water content that occur during MEA fabrication, and are therefore very difficult to handle during the MEA fabrication process.

In a further embodiment, a laminated membrane comprising more than one polymer-containing layer is provided, at least one layer of which is a composite membrane of the invention. Where a laminated membrane is formed that comprises more than one composite membrane layer of the invention, each layer may comprise either the same or different types of fibres and porous substrates, and also the same or different types of polymeric material embedded within the porous substrate of each composite membrane layer. Using such a laminated structure, it is possible, for example, to tailor the properties of the laminate membrane opposed to the anode and cathode sides in the MEA of a proton exchange membrane fuel cell, for example, to improve water management in the fuel cell, or to be able to use lower cost proton-conducting polymers to form a substantial part of the laminate membrane.

Composite membranes comprising the substrate of the present invention are suitable for low cost manufacture, and the substrates and membranes may be manufactured by:

(i) forming a porous substrate of, preferably randomly orientated individual, mixed amorphous silica fibres by adapting a continuous manufacturing process, which for example may be based on wet lay processes such as those employed in paper-making, or dry lay processes employed, for example, to produce non-woven fabrics and felts; and, optionally, thereafter,

(ii) impregnating the fibre matrix substrate with the polymeric material to produce a membrane. This can be done by any number of coating processes such as printing, rolling, K-bar, doctor blade methods, spraying or thin-film casting.

For example, in a process based on a paper-making technology to prepare a composite membrane, the fibres are dispersed in water to form a dilute slurry and thereafter a continuous structure is formed by the controlled deposition of said slurry onto a moving mesh bed, de-watering the solids, and drying and compacting the fibre network. The solution containing the dispersion of the binder material can be applied either at the wet end of the process, *i.e.* before the drying stage, or after the network has been dried. This is followed by nip roller coating of the substrate to fill it with a solution of the ion-conducting polymeric material, and further compaction and drying of the membrane under a suitable time, temperature and pressure regime to produce the final thin film or sheet of fibre/polymer composite membrane.

A major advantage of using a continuous manufacturing method, such as a conventional paper making technique, is that the composite membrane is easily manufactured in a fewer number of steps than prior art composite membranes, thus making it more cost-effective and commercially viable. The membrane may also be produced in continuous lengths of many metres and widths of equal to or greater than one metre. A further advantage is that it is possible to combine a membrane of the present invention with one or more electrode layers as described in European patent specification number EP 0 791 974 to form a membrane electrode assembly at the same rate as each individual component could be produced.

The present invention also relates to a membrane electrode assembly and a method for the manufacture thereof, wherein the composite membrane is one according to the present invention. A still further aspect of the present invention relates to a fuel cell and a method for the manufacture thereof, which fuel cell comprises a composite membrane of the present invention.

The present invention is not limited to the use of the composite membrane in a fuel cell and any electrochemical device which comprises a composite membrane of the invention is within the scope.

The present invention will now be described by way of example only which is not intended to be limiting thereof.

EXAMPLE 1

PREPARATION OF MIXED AMORPHOUS SILICA/ALCOHOLIC NAFION® SUBSTRATE

A mixture of chopped silica fibres (Type QC9/33-20mm from Quartz et Silice BP 521-77794 Nemours, Cedex, France) (0.18g), and silica microfibre (Q fibre, type 106 from Johns Manville, Insulation Group, PO Box 5108, Denver, CO, USA) (0.37g) were dispersed with mixing, in water (3000ml). A non-woven matrix was fabricated from the resulting mixture in a single-step process, based on the principles of paper-making technology, as a sheet size of 855cm^2 (33cm diameter) in a sheet former (design based on standard SCA Sheet former from AB Lorentzen & Wettre, Box 4, S-163 93 Stockholm, Sweden). The fibre sheet was removed from the wire and air dried at 150°C.

The non-woven sheet was sprayed with a 5% solution of Nafion®, 1100 EW in lower aliphatic alcohols (Solutions Technologies Inc, Mendenhall, PA 19357, USA) to give a dry Nafion® loading of 0.78g.

EXAMPLE 2

PREPARATION OF MIXED AMORPHOUS SILICA/PTFE/SILICA SUBSTRATE

A non-woven matrix was fabricated according to the method and materials of Example 1. The fibre sheet, as formed on the wire and whilst still wet, was sprayed with a binder solution comprising a 10wt% aqueous dispersion of polytetrafluoroethylene (Teflon GP1®; ICI Chemicals and Polymers Ltd, PO Box 4, Thornton, Cleveleys, Blackpool, FY5 4QD) and a 10wt% solution of colloidal silica (Syton® T40AS; DuPont Speciality Chemicals, Havennummer 500, Wilmington Straat, 2030 Antwerp, Belgium) in a 1:1 ratio to give a loading of 0.27g of the Teflon/silica mixture. The sheet was removed from the wire and air dried at 150°C, then fired in air at 280°C

EXAMPLE 3

PREPARATION OF MIXED AMORPHOUS SILICA/SILICA SUBSTRATE

A non-woven matrix was fabricated according to the method and materials of Example 1. The fibre sheet, as formed on the wire and whilst still wet, was sprayed with a binder solution comprising a 20wt% solution of colloidal silica (Syton® T40AS; DuPont Speciality Chemicals, Havennummer 500, Wilmington Straat, 2030 Antwerp, Belgium) to give a loading of 0.1g of the silica. The sheet was removed from the wire and air dried at 150°C.

COMPARATIVE EXAMPLES

NAFION® 1135, 115 & 117 MEMBRANES

Nafion® membrane type 1135 (produced by E I DuPont de Nemours, Polymer Products Department, Fayetteville, NC, USA) was used as received. A 10x10cm square was cut from the bulk membrane. A measurement of the membrane's mass was taken before the sample was placed in a sealable polyethylene bag of known weight. With the bag seal open, the membrane was dried overnight (~16 h) at 40°C under vacuum (~10mbar). After releasing the vacuum, the bag was quickly sealed before being weighed. [Mass loss from the membrane and bag together was adjusted for the average mass loss from three identical bags containing no membrane]. Lengths in the x and y directions were measured whilst the dried membrane was still in the sealed bag to establish the dehydrated dimensions.

The membrane was placed in 2 litres of de-ionised water, heated to boiling and maintained at boiling for 90 minutes. The membrane was then removed from the de-ionised water and the excess surface water removed by blotting with filter paper. The x and y dimensions were then measured using the same procedure as before.

Nafion® membranes types 115 and 117 (also produced by E I DuPont de Nemours, Polymer Products Department, Fayetteville, NC, USA) were also used as received. A 10x10cm square was cut from each bulk membrane and treated according to the above procedure.

The dimensional changes and area change for each comparative membrane are recorded in Table 1.

EXAMPLE 4

PREPARATION OF TRIPLE LAMINATE MEMBRANES USING SUBSTRATE OF EXAMPLE 1

The non-woven silica fibre/binder matrix prepared according to Example 1 was placed on a sheet of sintered PTFE and a solution of perfluorosulphonic acid (Nafion® produced by E I DuPont de Nemours) in the aqueous form as described in EP 731 520 was applied to the silica fibre matrix. The structure was filled with the aqueous Nafion® to achieve a total solid Nafion® loading of 6.55mg/cm^2.

A further two sheets were prepared in the same fashion. The three sheets were placed on top of each other and sandwiched between two thin, non-porous PTFE sheets. The sandwich was pressed at 90 to 100psig for six minutes at 177°C to produce a triple laminate membrane.

A 10x10cm square was cut from the bulk membrane and treated by the same procedure as described in the Comparative Examples. The results are recorded in Table 1.

EXAMPLE 5

PREPARATION OF TRIPLE LAMINATE MEMBRANES USING SUBSTRATE OF EXAMPLE 2

The non-woven silica fibre/binder matrix prepared according to Example 2 was treated according to the method and materials of Example 4 (total solid Nafion® loading of 7.3mg/cm^2) to produce a triple laminate membrane, whose results also appear in Table 1.

EXAMPLE 6

PREPARATION OF TRIPLE LAMINATE MEMBRANES USING SUBSTRATE OF EXAMPLE 3

The non-woven silica fibre/binder matrix prepared according to Example 3 was treated according to the method and materials of Example 4 (total solid Nafion® loading of 7.29mg/cm^2) to produce a triple laminate membrane, whose results also appear in Table 1.

EXAMPLE 7

PREPARATION OF SINGLE SHEET MEMBRANES USING SUBSTRATE OF EXAMPLE 1

A single sheet of the non-woven mixed silica fibre matrix with the sprayed alcoholic Nafion® binder was formed as described in Example 1 and filled with a solution of perfluorosulphonic acid (Nafion® produced by E I DuPont de Nemours) in the aqueous form as described in EP 731 520 to achieve a total solid Nafion® loading of 6.49mg/cm^2.

The sheet was sandwiched between two thin, non-porous PTFE sheets. The sandwich was pressed at 90 to 100psig for six minutes at 177°C to produce a membrane.

A 10x10cm square was cut from the bulk membrane and treated by the same procedure as described in the Comparative Examples. The results are recorded in Table 1.

EXAMPLE 8

PREPARATION OF SINGLE SHEET MEMBRANES USING SUBSTRATE OF EXAMPLE 2

The non-woven silica fibre/binder matrix prepared according to Example 2 was treated according to the method and materials of Example 7 (total solid Nafion® loading of 7.24mg/cm^2) to produce a membrane whose results also appear in Table 1.

EXAMPLE 9

PREPARATION OF SINGLE SHEET MEMBRANES USING SUBSTRATE OF EXAMPLE 3

The non-woven silica fibre/binder matrix prepared according to Example 2 was treated according to the method and materials of Example 7 (total solid Nafion® loading of 6.38mg/cm^2) to produce a membrane whose results also appear in Table 1.

TABLE 1

SILICA MIXED FIBRE MEMBRANES

Example Number	Membrane	Binder Type	Post-boil Dimensional Changes		
			x (%)	y (%)	Area (%)
CP	Nafion® 1135	N/A	+4.1	+25.0	+30.0
CP	Nafion® 115	N/A	+15.8	+20.5	+39.0
CP	Nafion® 117	N/A	+13.4	+22.5	+39.0
4	triple laminate	alcoholic Nafion®	+0.5	0	+0.5
5	triple laminate	1:1 colloidal silica/PTFE	+1.5	+3.0	+4.6
6	triple laminate	colloidal silica	+3.0	+2.0	+5.6
7	single sheet	alcoholic Nafion®	-1.0	-1.0	-2.0
8	single sheet	1:1 colloidal silica/PTFE	-4.0	-4.5	-8.0
9	single sheet	colloidal silica	-1.0	-2.0	-3.0

CLAIMS

1. A substrate, suitable for the preparation of a composite membrane, which substrate comprises a porous matrix of fibres, characterised in that the fibres comprise mixed amorphous silica fibres that are bound with a binder.

2. A substrate according to claim 1, wherein the mixed amorphous silica fibres comprise micro-fine amorphous silica fibres.

3. A substrate according to claim 1 or claim 2, wherein the mixed amorphous silica fibres comprise one or more chopped strand(s) of amorphous silica.

4. A substrate according to any preceding claim wherein the amorphous silica fibres comprise a mixture of both microfibres and chopped fibres in the range of from 95:5% to 5:95% by weight of the mixture respectively.

5. A substrate according to claim 4 wherein the amorphous silica fibres comprise a mixture of both microfibres and chopped fibres in the range of from 70:30% to 30:70% by weight of the mixture respectively.

6. A substrate according to any preceding claim wherein the fibres have a diameter in the range of from 0.1μm to 50μm.

7. A substrate according to claim 6 wherein the fibres have a diameter in the range of from 0.4μm to 9μm.

8. A substrate according to any preceding claim, wherein the binder comprises a solution or dispersion of ion-exchange polymeric materials, or non-ion-conducting polymers, or inorganic materials or mixtures thereof.

9. A substrate according to any preceding claim for use in the preparation of a composite membrane.

10. A composite membrane comprising a porous substrate of fibres and at least one ion-conducting polymer, characterised in that the substrate is one according to any preceding claim, which comprises mixed amorphous silica fibres bound with a binder.

11. A composite membrane according to claim 10, which when tested by the method described herein in the Examples, results in less than or equal to about ±9% change in the area.

12. A composite membrane according to claim 10 or claim 11 wherein the total thickness of the membrane is less than 200μm.

13. A composite membrane according to any one of claims 10 to 12 for use in a fuel cell.

14. A process for the manufacture of a substrate according to any one of claims 1 to 9, which process comprises

(a) dispersing the fibres in water to form a slurry;

(b) depositing the slurry onto a mesh bed to form a network;

(c) drying and compacting the fibre network; and

(d) applying, before or after step (c), a dispersion of binder.

15. A process for the manufacture of a membrane according to any one of claims 10 to 13, which process comprises

(i) forming a porous substrate of, preferably randomly orientated individual mixed amorphous silica fibres bound with a binder by a process according to claim 14; and, thereafter,

(ii) impregnating the porous substrate with a polymeric material to produce a membrane.

16. A process according to claim 15, wherein step (ii) is carried out by nip roller coating of the substrate to fill it with a solution of ion-conducting polymeric material, and further compaction and drying of the membrane.

17. A membrane electrode assembly comprising a substrate according to any one of claim 1 to 9 and/or a composite membrane according to any one of claims 10 to 13.

18. A fuel cell comprising a substrate according to any one of claim 1 to 9 and/or a composite membrane according to any one of claims 10 to 13.

INTERNATIONAL SEARCH REPORT

Interr nal Application No
PCT/GB 99/02935

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C08J5/22 H01M8/10

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C08J H01M B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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A	PATENT ABSTRACTS OF JAPAN vol. 006, no. 266 (C-142), 25 December 1982 (1982-12-25) & JP 57 159502 A (TOYO BOSEKI KK), 1 October 1982 (1982-10-01) abstract	1

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[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

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"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

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"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
16 December 1999	12/01/2000

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A	US 5 523 181 A (STONEHART PAUL ET AL) 4 June 1996 (1996-06-04) claims 1,3,5	1
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